

REMARKS

Claims 1-3, 7-14 and newly added claims 15 and 16 are pending in this application. Claims 9 and 10 have been canceled, without prejudice, as being redundant in view of the replacement thereof by the addition of independent claims 15 and 16.

The examiner's attention is drawn to the preliminary amendment, which was filed concurrently with the Request for Continued Examination on June 17, 2002. In the preliminary amendment, claim 1 has been amended to provide for the amount of surface active material present in the graphite material, and a range of 0.1 to 10.0% by weight based upon the weight of graphite material has been designated for the amount of surface active material. Enclosed with this response is a Declaration of Katsutomo Ozheki, the inventor of the present application, who has been engaged in the research and development of electrodes for secondary cells for a period of at least 12 years. Experiments have been carried out by Mr. Ohzeki to show that the range of 0.01 to 10.0 wt. % of surface active material on the conductive graphite is a critical range and is associated with an improved performance on the part of the so-treated graphite in graphite powder for negative electrodes used in lithium ion secondary cells. As stated in the Results and Conclusions on page 5 of Mr. Ozheki's Declaration, outside of the range of 0.01 to 10 wt. %, the surface active material of less than 0.01 wt. %

resulted in discharge and charge capacities which were large and Coulomb efficiencies were small. The discharge capacities increased within the range of up to 10.0 wt. % and the difference in discharge capacities were small. The cited Japanese Publication 09147916 (JP '916) does not disclose the use of starch or polysaccharides for the surface active layer on the graphite. The JP '916 reference, moreover, does not indicate that the protective layer formed on the solid particles is of critical quantity nor that there is anything to be gained by controlling the amount of protective layer.

In the JP '916 reference, in the negative electrode and/or the positive electrode, at least one protecting layer is formed. The protective layer is formed of solid particles (inorganic chalcogenide particles) and a water soluble polymer (polyacrylic derivative or a cellulose derivative). This means that "a negative electrode, the surface of which is coated with only one protective layer" is shown in the cited reference.

In the present invention, graphite particles are forming a negative electrode, and each graphite particle has adsorbed or is coated with a layer of surface active material. In other words, "a specific quantity of water soluble polymeric material is applied to the innumerable number of surfaces of graphite particles." In addition, in order to form the protective layer,

"the solid particle" are not necessary, which is different from the cited reference.

Accordingly, the state of formation of protective layer on the surfaces of "the respective component particles" in the present invention is quite different from the state of formation of protective layer in the cited reference, that is, the protective layer is formed on "the mass of electrode itself."

Furthermore, the quantity of the coating of the surface active material on graphite is critical and the limit of 0.01 to 10.0 wt. % of the surface active polymer is neither disclosed nor suggested in the reference.

Still further, in the cited reference, conductive particles such as the carbonaceous compound, natural graphite and vapor phase growth carbon are contained in the protective layer because the protective layer is as thick as 1 to 4  $\mu\text{m}$ . On the other hand, in the present invention, each particle of the electrode material is treated by the surface active material by using neither the solid particles nor the conductive material

In view of the above comments together with the enclosed Declaration, reconsideration and allowance of the present application is respectfully solicited.

Should there be any outstanding matters that need to be resolved in the present application, the Examiner is respectfully requested to contact Edward H. Valance (Reg. No. 19,869) at the telephone number of the undersigned below, to conduct an interview in an effort to expedite prosecution in connection with the present application.

If necessary, the Commissioner is hereby authorized in this, concurrent, and future replies, to charge payment or credit any overpayment to Deposit Account No. 02-2448 for any additional fees required under 37 C.F.R. §§ 1.16 or 1.17; particularly, extension of time fees.

Respectfully submitted,

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Attachment: Declaration of Katsutomo Ohzeki

(Rev. 02/20/02)



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PATENT  
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IN THE U.S. PATENT AND TRADEMARK OFFICE

Applicant: OHZEKI, et al. Conf.: 6799  
Appl. No.: 09/254,316 Group: 1754  
Filed: March 4, 1999 Examiner: HENDRICKSON, S.  
For: GRAPHITE POWDER FOR NEGATIVE ELECTRODE  
OF LITHIUM ION SECONDARY CELL AND  
METHOD OF PRODUCING THE SAME

DECLARATION OF KATSUTOMO OHZEKI

Assistant Commissioner for Patents  
Washington, DC 20231

Sir:

1. The undersigned Declarant is the inventor of Application Serial No. 09/254,316 filed on March 4, 1999.

2. Declarant is a graduate of Tokai University, Graduate School of Science, Course of Chemistry with a degree in Master of Chemistry granted in 1990. Declarant has been employed by Hitachi Powdered Metals Co., Ltd. for 12 years and is engaged in research and development of electrodes for secondary cells, including lithium ion cells for batteries.

3. Declarant is familiar with the prosecution of Application Serial No. 09/254,316 filed on March 4, 1999, and the officially cited prior art, Japanese Patent Publication No. 09147916 (hereinafter known as "JP '916").

4. Declarant has caused certain experiments to be carried out under his direction as described hereinbelow:

4(a). Experiment 1

A predetermined quantity of water-soluble cellulose of hydroxyethyl cellulose was dissolved into 1 liter of pure water and 100 g of graphitized mesophase carbon micro-beads having an average particle diameter of 6  $\mu\text{m}$  were added thereto. The resulting solution was stirred for 60 minutes with a propeller stirrer so as to disperse the contents. The thus treated liquid was dried with a spray drier to obtain a dry powder that was evaluated with the following results.

The determined quantities of adsorbed/coated hydroxyethyl cellulose in the treated powders and the results of evaluation of the use of treated powders are shown in the following Table 1. Also in the cases of the carbon material of graphitized mesophase carbon microbeads, when the adsorbed/coated quantity of surface active effect material of hydroxyethyl cellulose was less than 0.01 wt. % (Sample Nos. 1.1, 1.2), the ratios of charge capacities and discharge capacities were large (Coulomb efficiencies were small). On the other hand, when the quantities of adsorbed/coated hydroxyethyl cellulose were in the range of 0.01 to 10 wt. % (Sample Nos. 1.3-1.6), the discharge capacities increased and the differences in discharge capacities were small. Thus, the improvement in the characteristics of the cells was seen when the content of hydroxyethyl cellulose was in the range of 0.01 to 10.0 gms.

Table 1

Sample Number	Content of Hydroxyethyl Cellulose (HEC) (g)	Adsorbing /Coating Qty. (wt.%)	Capacity (mAh/g)		Coulomb Efficiency (%)	Capacity Retention Rate (%)
			Charge	Discharge		
1.1	- - -	- - -	286	240	84	75
1.2	0.01	0.009	295	242	82	75
1.3	0.02	0.018	281	250	89	82
1.4	1.5	1.4	287	267	93	86
1.5	7.0	6.8	287	270	94	88
1.6	10.0	9.9	287	270	94	88
1.7	15.0	14.7	246	219	89	86

#### 4(b). Experiment 2

A predetermined quantity of 1:1 mixture of a starch derivative of acetate starch and water-soluble synthetic acrylic resin was dissolved into 1 lit. of pure water and 100 g of graphitized pitch coke was added thereto. The resultant solution was stirred for 120 minutes with a homogenizer so as to disperse the contents. The thus treated liquid was filtered with #5B filter paper to separate the treated powder from the medium. The treated powder was dried with a spray drier to obtain dry powder which was evaluated as follows.

The determined quantities of adsorbed/coated surface active effect materials in the treated powders and the results of

evaluation in the use of treated powders are shown in the following Table 2. Also in the cases of the carbon material of pitch coke, when the adsorbed/coated quantity of surface active effect material was less than 0.01 wt. % (Sample Nos. 2.1,2.2), the ratios of discharge capacities and charge capacities were large (Coulomb efficiencies were small). On the other hand, when the quantities of adsorbed/coated surface active effect material are in the range of 0.01 to 10 wt. % (Sample Nos. 2.3-2.6), the discharge capacities increased and the difference in discharge capacities were small.

Table 2

Sample Number	Contents of Acetate Starch +Acrylic Resin (g)	Adsorbing /Coating Qty. (wt.%)	Capacity (mAh/g)		Coulomb Efficiency (%)	Capacity Retention Rate (%)
			Charge	Discharge		
2.1	- - -	- - -	340	250	74	78
2.2	0.01	0.009	338	250	74	79
2.3	0.02	0.018	350	280	80	89
2.4	1.5	1.4	366	300	82	89
2.5	7.0	6.8	351	305	87	90
2.6	10.0	9.9	341	300	88	88
2.7	15.0	14.7	235	200	85	86



5. Results and Conclusions

Experiments 1 and 2 are self-explanatory. Without any chalcogenide particles, but otherwise analogous to the conductive particles of graphite as shown in Japanese Patent Publication No. 09147916, the critical range of the coating layer on the graphite particles is shown to be from 0.01 to 10 weight % (Sample Nos. 1.3-1.6 and also Sample Nos. 2.3-2.6). Outside of this range, when the surface active material was less than 0.01% weight, the discharge and charge capacities were large and Coulomb efficiencies were small. Within the range of 0.01-10% by weight of surface active material on conductive graphite, the discharge capacities increased and the difference in discharge capacities were small.

6. DECLARANT hereby states that all statements made herein of his own knowledge, and information and belief, are believed to be true; and, further, that these statements were made with the knowledge that willful, false statements, and the like, so made are punishable by fine or imprisonment or both under Section 1001 of Title 18 of the United States Code, and that such willful, false statements may jeopardize the validity of the application, any patent issuing thereon or any patent to which this verified statement is direct.

Date: Oct. 24, 2002

Katsunomo Ohzeki, Declarant